



*“DSRC Nanoscopic Phonon Engineering Workshop”*

*Booz Allen Hamilton, Arlington, May 9, 2005*



**Phonon-Polariton Physics:**  
**Thermal Conductivity, Phonon-Polariton Lasers**  
**and**  
**Phonon Transistors in Nanostructures**

**Anvar Zakhidov on behalf of UTD PEOM Team**

*NanoTech Institute*  
*University of Texas at Dallas*

Anvar A. Zakhidov, University of Texas at Dallas



**Outline :**

**Motivation for Tuning Phonon  $K(T)$  in Our 2 Main Systems.**

**System 1.**

**Phonon-Polaritons: New Mechanism of Thermal Conductivity**  
**Phonon-Polariton Lasers**

**Overview of Phonon-Polaritonics**

**System 2.**

**Carbon Nanotube Yarns and Sheets for Enhanced Thermal Conductivity**  
**Phonon Transistor with Charge Injection Gate**

Anvar A. Zakhidov, University of Texas at Dallas

Report Documentation Page			Form Approved OMB No. 0704-0188		
Public reporting burden for the collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to a penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number.					
1. REPORT DATE <b>09 MAY 2005</b>		2. REPORT TYPE <b>N/A</b>		3. DATES COVERED <b>-</b>	
4. TITLE AND SUBTITLE <b>Phonon-Polariton Physics: Thermal Conductivity, Phonon-Polariton Lasers and Phonon Transistors in Nanostructures</b>				5a. CONTRACT NUMBER	
				5b. GRANT NUMBER	
				5c. PROGRAM ELEMENT NUMBER	
6. AUTHOR(S)				5d. PROJECT NUMBER	
				5e. TASK NUMBER	
				5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) <b>NanoTech Institute University of Texas at Dallas</b>				8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)				10. SPONSOR/MONITOR'S ACRONYM(S)	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S)	
12. DISTRIBUTION/AVAILABILITY STATEMENT <b>Approved for public release, distribution unlimited</b>					
13. SUPPLEMENTARY NOTES <b>See also ADM001801., The original document contains color images.</b>					
14. ABSTRACT					
15. SUBJECT TERMS					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT <b>UU</b>	18. NUMBER OF PAGES <b>33</b>	19a. NAME OF RESPONSIBLE PERSON
a. REPORT <b>unclassified</b>	b. ABSTRACT <b>unclassified</b>	c. THIS PAGE <b>unclassified</b>			



## Our Main Systems and Materials:



### 1. New Mechanism of Phonon-Polariton Thermal Conductivity

### 2. Carbon Nanotubes with Enhanced $K(T)$ CNT in CNT Yarns and Oriented CNT bucky-aerogels

Anvar A. Zakhidov, University of Texas at Dallas



## Our Main Concepts: “Solid State Heat Pipes” And “Phonon Transistor or Valve”



Two types of heat pipes:

1. We predict an analog of optical fiber for heat transfer by light mixed with optical phonon

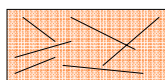
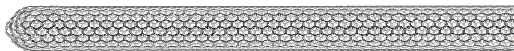


Wave-guide in which Phonon-Polariton can propagate like light and contribute to  $K(T)$  of low-K organic matter

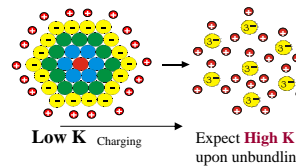
2. CNT are nanoscale analog of optical fibers in which phonons flow is ballistic and 1-d. Can it be a Heat-pipe

How to preserve this ballistic transport and high  $K$  in fibers of CNTs and CNT dispersed in polymers?

Giant  $K(T) = 3000 - 6000$  W/mK,  
Predicted in SWCNT and measured in MWCNTs



a) Conjugated Polymers-CNT for distributed heat removal in Organic Electronics



Anvar A. Zakhidov, University of Texas at Dallas



## Physical Ideas for Engineering Phonons at Nanoscale for Enhanced thermal conductivity K(T)



1. Optical Phonons dispersion is flat, group velocity small: no contribution to K(T)
2. It can be really engineered: changed to **Phonon-Polariton with large Vg** and we found sizable contribute to thermal conductivity K(T).
3. In Organic matter a **Phonon-Laser** is proposed, which can be pumped by heat and produce monochromatic and coherent IR
4. In single Carbon nanotubes phonons are ballistic and  $L_m \sim 1 \mu m$ , providing high K(T) for one CNT.  
Our Approach is to achieve and control high K(T) in real systems:
  - unbundle tubes: **decrease Phonon-Phonon scattering both inter-tube and UP**
  - coat tubes with polymers and mix into a matrix of low-K conjugated polymer at concentration lower or close to percolation for nano-scale distributed heat removal.
5. **Phonon-Transistor concept** appeared as a result of tunable K(T)

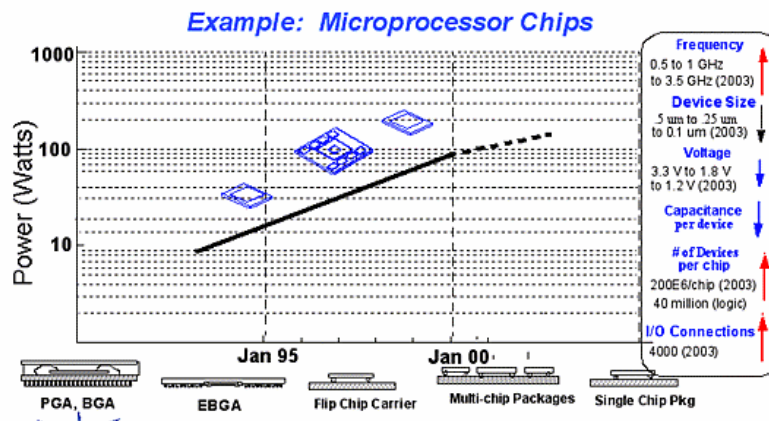
Anvar A. Zakhidov, University of Texas at Dallas



## DoD Needs Better Cooling Systems for Electronic Warfare Chips



### High-performance Chip Dissipation



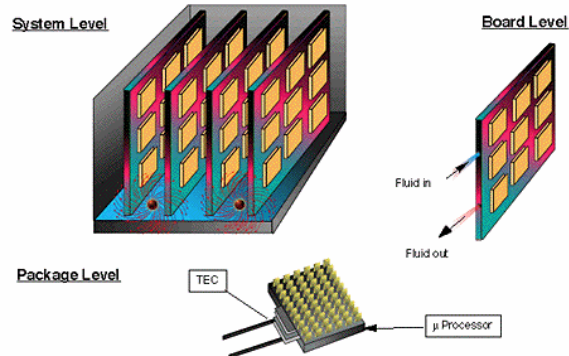
Anvar A. Zakhidov, University of Texas at Dallas



DARPA has a strong interest in Thermal Management: Previous HERETIC Program and Phonon Engineering Program are examples



Conventional Thermal Management Hierarchy



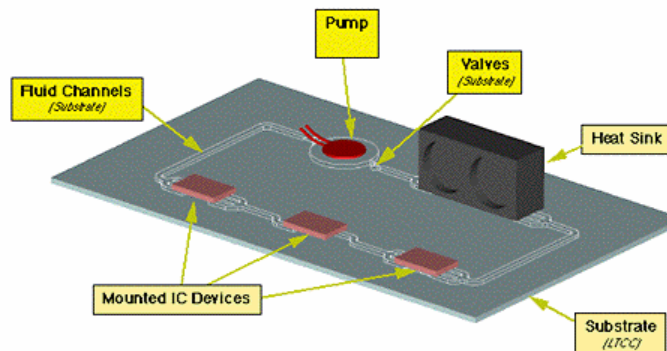
Anvar A. Zakhidov, University of Texas at Dallas



State-of-Art Heat Removal Circuitry Targets Fluids: Solid State Systems for cooling would be much more attractive for DoD Applications



Fluidic Based Cooling Using Channels



Anvar A. Zakhidov, University of Texas at Dallas



## Impact 1: for Heat Management Circuitry for DoD Applications



### Goal:

Develop micro- and nano-scale solid state heat removal circuitry with control devices (valves, couplers, switches, etc.) which can be ultimately integrated with electronic and photonic circuitry. This technology will be unique, since it will enable controllable, adaptable, distributed and programmable thermal management

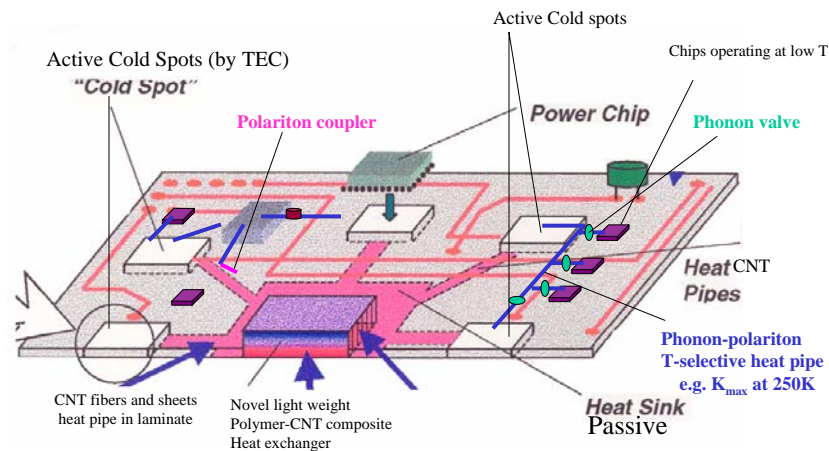
### Impact:

Efficient, compact and controllable (with active feedback) heat removal circuitry will enable design of low power, small form-factor DoD systems, such as radars, high performance computers, and other electronic and photonic warfare subsystems

Anvar A. Zakhidov, University of Texas at Dallas



## Our Concept: Solid State Cooling Circuitry Integrated on Chip, Heat Pipes (CNTs or "Polaritonic Fibers")



Anvar A. Zakhidov, University of Texas at Dallas

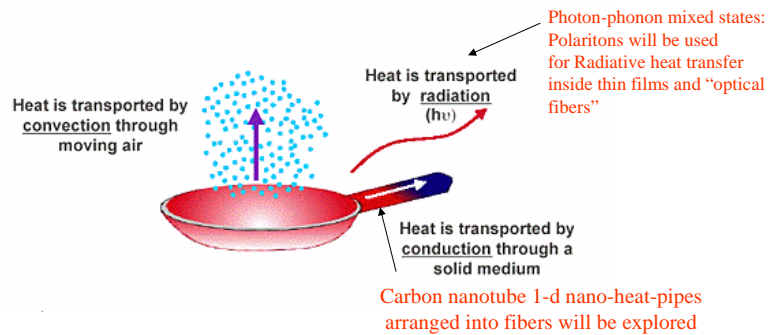


## Physical Concepts for Solid State Heat Pipes:

1. Heat Transfer by Polaritons in Organic Thin Films
2. Achieve high  $K(T)$  of Carbon Nanotubes with engineered Phonon-phonon interaction



### Modes of Heat Transport



Anvar A. Zakhidov, University of Texas at Dallas



## Phonon-Polaritons Contribution to Thermal Conductivity

Prospects for Organic Phonon-Polariton  
Heat Pipes  
And Phonon-Polariton Laser

Overview of Polaritonics (MIT, France,)

Anvar A. Zakhidov, University of Texas at Dallas



## Thermal conductivity in solis:



- Known: by phonons, photons, electrons
  - **New: by Phonon-Polaritons**
- Bulk and Thin Films (Microcavity)
  - Ballistic regime:  $\Lambda(\omega) > d$
  - Statistical, diffusive regime  $\Lambda(\omega) \ll d$
- Phonon-Polariton Microcavity Laser

Anvar A. Zakhidov, University of Texas at Dallas



## Motivation



- Contribution of phonon-polaritons to thermal conductivity **has never been estimated**  
(Klemens P.G. et al. Term. Conduct. (1988), 19, 453 )
- Phonon-polaritons can have **very long** mean free paths  $\Lambda(\omega)$  of mm and even cm – **ballistic propagation**
- **Enhancement** of  $K(T)$  may be expected in nanostructures with sizes,  $d$  smaller than  $\Lambda(\omega)$

In thick samples  $K(T)$  still can have contribution from polaritons

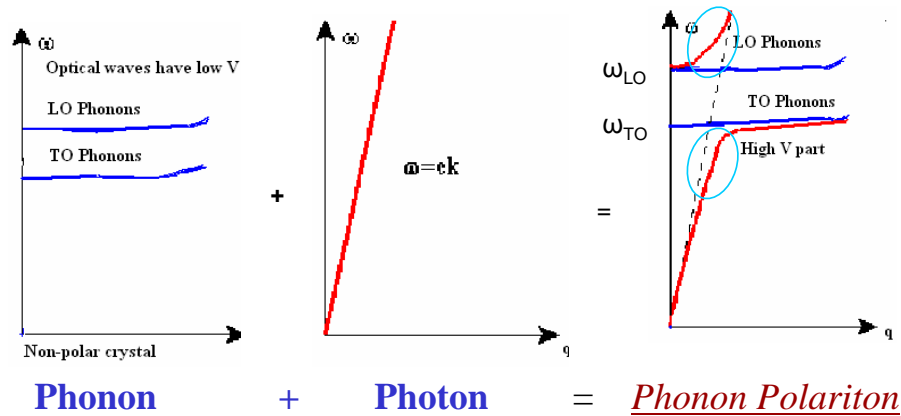
Anvar A. Zakhidov, University of Texas at Dallas



## What is phonon-polariton?



Polariton is a mixed excitation, has phonon and electromagnetic components, and can propagate with high velocity



Anvar A. Zakhidov, University of Texas at Dallas

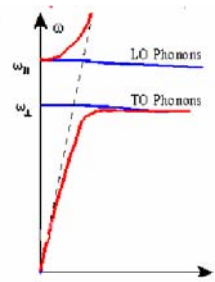
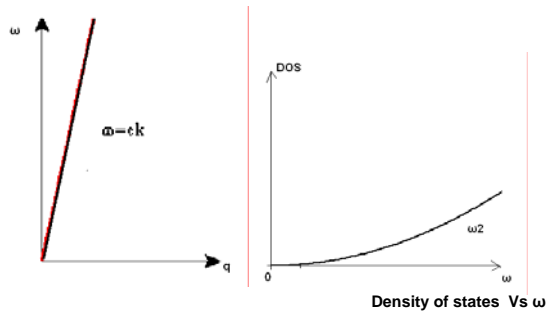


## POLARITONS vs PHOTONS



Photons in the Bulk,  $\epsilon = \epsilon_\infty$  - constant

Polaritons: Coherent mixture of Photon with Phonon



For Transverse Waves:

$$q^2 \epsilon^2 / \omega^2 = \epsilon_\infty$$

$$\omega = q \epsilon / \sqrt{\epsilon_\infty}$$

Density of States:

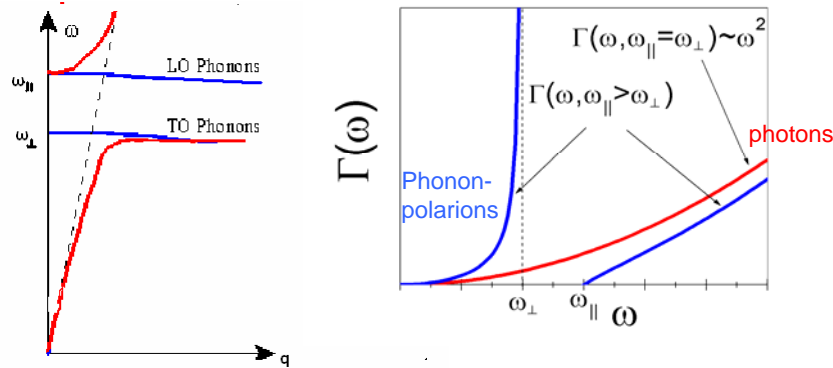
$$\text{DOS} \sim q^2 dq / (2\pi)^3 \sim \omega^2 d\omega$$

DOS becomes singular at  $\omega_T$

Anvar A. Zakhidov, University of Texas at Dallas



## Phonon-Polaritons in Bulk: Dispersion and DOS



Anvar A. Zakhidov, University of Texas at Dallas

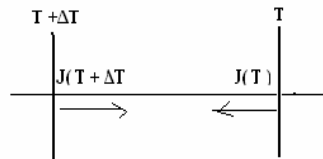


## PHOTONS: Radiative Contribution to Thermal Conductivity in ballistic regime (Landauer)

$$J_z = 2 \int \left( \frac{k^2}{(2\pi)^3} \right) \left( \frac{1}{e^{\hbar\omega(k)/k_B T} - 1} \right) (\hbar\omega(k)) \left( \frac{d\omega}{dk} \right) dk \sin \theta \cos \theta d\phi$$

Total current  $J = J_z(T + \Delta T) - J_z(T)$

$$\frac{J(T)}{\Delta T} \cong 26 \frac{k_B c}{(2\pi)^2 \sqrt{\epsilon_{\infty}}} \left( \frac{k_B T \sqrt{\epsilon_{\infty}}}{\hbar c} \right)^3$$



Anvar A. Zakhidov, University of Texas at Dallas



## Ballistic Propagation



$$\mathcal{E} = \mathcal{E}_\infty$$

$$J(T) = 2 \int_0^\infty \frac{q^2 dq \sin \theta}{(2\pi)^3} \cos \theta \hbar \omega \frac{d\omega}{dq} [\eta_{hot} - \eta_{cold}]$$

$$\frac{J(T)}{\Delta T} = \frac{k_B c}{(2\pi)^2 \sqrt{\mathcal{E}_\infty}} \left( \frac{k_B T \sqrt{\mathcal{E}_\infty}}{\hbar c} \right)^3 \int_0^\infty \frac{x^4 e^x}{(e^x - 1)^2} dx$$

But we have,  $\int_0^\infty \frac{x^4 e^x}{(e^x - 1)^2} dx \cong 26$

$$\frac{J(T)}{\Delta T} \cong 26 \frac{k_B c}{(2\pi)^2 \sqrt{\mathcal{E}_\infty}} \left( \frac{k_B T \sqrt{\mathcal{E}_\infty}}{\hbar c} \right)^3$$

Anvar A. Zakhidov, University of Texas at Dallas



## Phonon-Polaritons:

Contribution to Thermal Conductivity:  
Resonance with Optical Phonon at  $\omega = \omega_\perp$



$$k^2 = \frac{\mathcal{E}_\infty \omega^2}{c^2} \left( \frac{\omega_\parallel^2 - \omega^2}{\omega_\perp^2 - \omega^2} \right), \quad \mathcal{E}(\omega) = \mathcal{E}_\infty \left( \frac{\omega_\parallel^2 - \omega^2}{\omega_\perp^2 - \omega^2} \right)$$

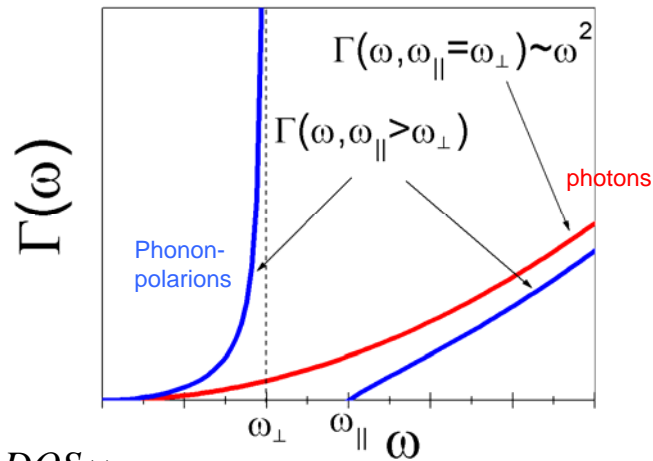
$$\begin{aligned} \frac{J^{BULK}(T)}{\Delta T} = & \frac{k_B c}{(2\pi)^2 \sqrt{\mathcal{E}_\infty}} \left( \frac{k_B T \sqrt{\mathcal{E}_\infty}}{\hbar c} \right)^3 \left\{ \int_0^{x_\perp} \left( \frac{(x_\parallel^2 - x^2)(x_\perp^2 - x^2)}{(x_\perp^2 - x^2)^2 + 4x^2 x_\perp^2 \delta^2} \right) \frac{x^4 e^x}{(e^x - 1)^2} dx + \right. \\ & \left. + \int_{x_\parallel}^\infty \left( \frac{(x_\parallel^2 - x^2)(x_\perp^2 - x^2)}{(x_\perp^2 - x^2)^2 + 4x^2 x_\perp^2 \delta^2} \right) \frac{x^4 e^x}{(e^x - 1)^2} dx \right\} \end{aligned}$$

$$x_\perp = \frac{\hbar \omega_\perp}{k_B T}, \quad x_\parallel = \frac{\hbar \omega_\parallel}{k_B T}$$

Anvar A. Zakhidov, University of Texas at Dallas



## Major Difference: Density of States



$$\Gamma(\omega) \propto DOS \times v_{GROUP}$$

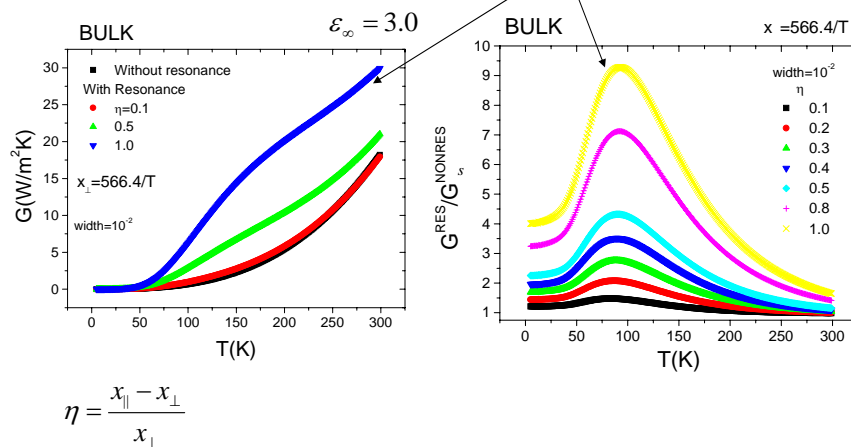
Anvar A. Zakhidov, University of Texas at Dallas



## Resulting Enhancement of Heat Conductance



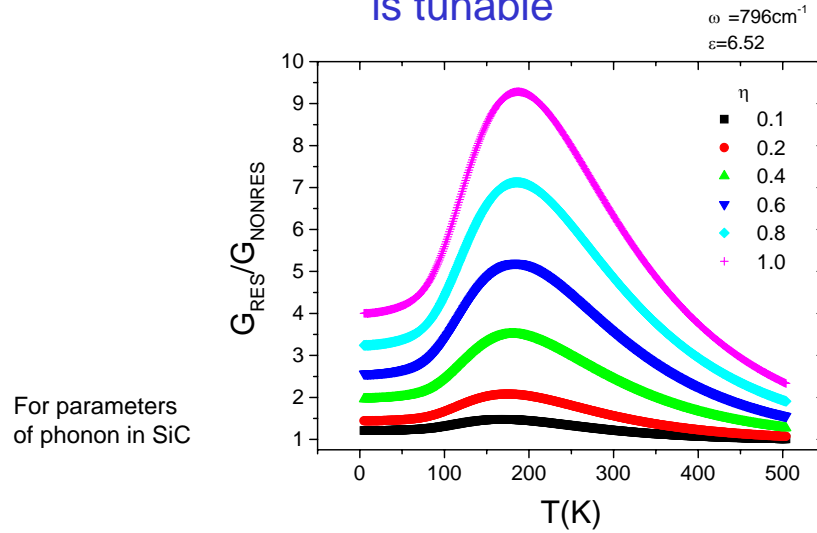
for parameters of MgO



Anvar A. Zakhidov, University of Texas at Dallas



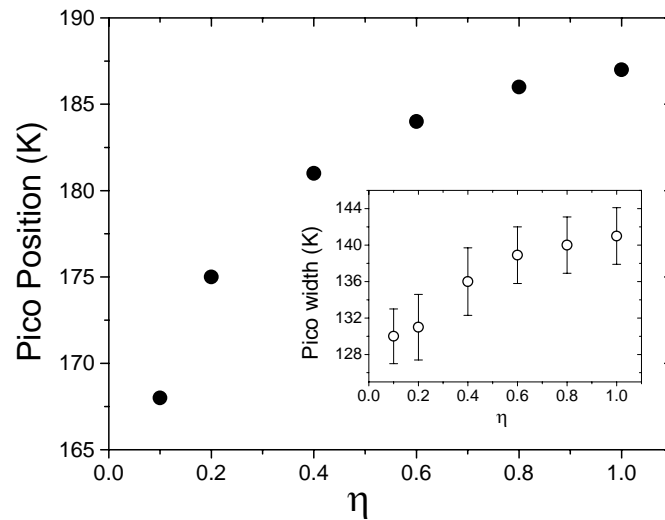
## Position of the enhancement peak is tunable



Anvar A. Zakhidov, University of Texas at Dallas



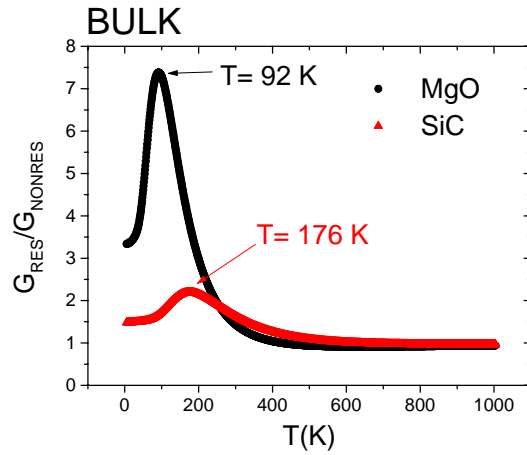
## The Dependence of T-Peak Position on the Tuning Parameter



Anvar A. Zakhidov, University of Texas at Dallas



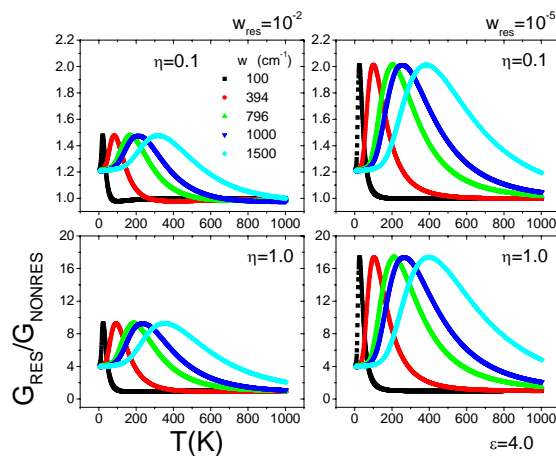
## Comparison of Polaritonic $K(T)$ in SiC and MgO



Anvar A. Zakhidov, University of Texas at Dallas



## Position and intensity of the enhancement peak is tunable



- Depends on:
- Frequency  $\omega_{\text{TO}}$
  - Line width  $\delta\omega$
  - Polaritonic gap  $\Delta$

To get a peak at RT, we need optical phonons with  $\omega_{\text{TO}} = 1500 \text{ cm}^{-1}$

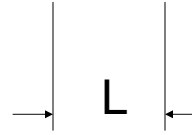
Anvar A. Zakhidov, University of Texas at Dallas



## Thin Films: Radiative contribution to thermoconductivity



$$q^2 = \frac{\omega^2 \varepsilon_\infty}{c^2} - \left( \frac{\pi s}{L} \right)^2$$



Film thickness

$$L = \lambda_{\text{TO}}/2$$

□

$$G^{\text{eff}}(L, T) = \frac{1}{L} \frac{J^{MC}}{\Delta T} = \frac{2}{L} \frac{k_B \varepsilon_\infty}{c(2\pi)^2} \left( \frac{k_B T}{\hbar} \right)^2 \sum_{s=1}^{\infty} F(x_s)$$

$$F(x_s) \equiv \int_{x_s}^{\infty} x^2 \sqrt{x^2 - x_s^2} \frac{e^x}{(e^x - 1)^2} dx$$

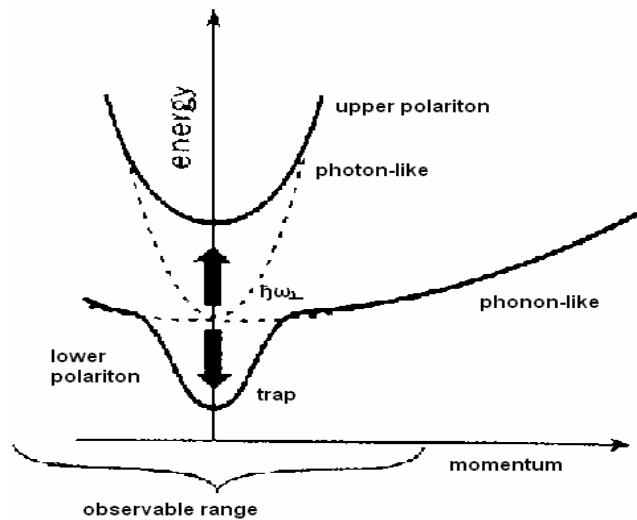
$$\omega(q=0) = \frac{\pi c}{L \sqrt{\varepsilon_\infty}}$$

$$x_s \equiv \frac{\hbar \omega(q=0)}{k_B T} s, \quad s = 1, 2, \dots$$

Anvar A. Zakhidov, University of Texas at Dallas



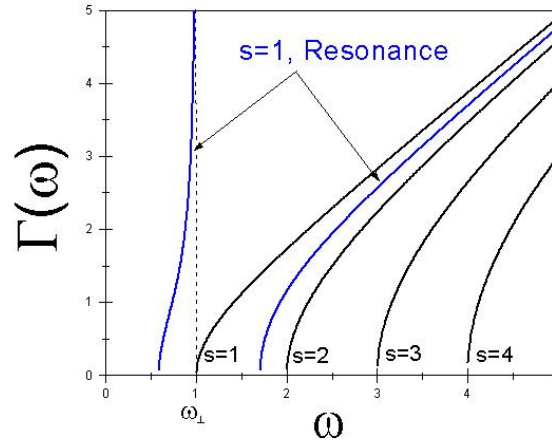
## Dispersion of Cavity polariton



Anvar A. Zakhidov, University of Texas at Dallas



$$\Gamma(\omega) \propto DOS_{MC} \times v_{GROUP}$$



Anvar A. Zakhidov, University of Texas at Dallas



**Thin Films:**  
Phonon-polariton contribution  
to thermal conductivity

$$q^2 = \frac{\varepsilon_\infty}{c^2} \left( \omega^2 - \omega_c^2 + \omega^2 \left( \frac{\omega_\perp^2 - \omega_\parallel^2}{\omega^2 - \omega_\perp^2} \right) \right)^2$$

$$\frac{J^{MC}}{\Delta T} = \frac{k_B \varepsilon_\infty}{c(2\pi)^2} \left( \frac{k_B T}{\hbar} \right)^2 \left\{ \int_{x_1}^{x_\perp} f(x, x_c, x_\parallel, x_\perp) dx + \int_{x_2}^{\infty} f(x, x_c, x_\parallel, x_\perp) dx \right\}$$

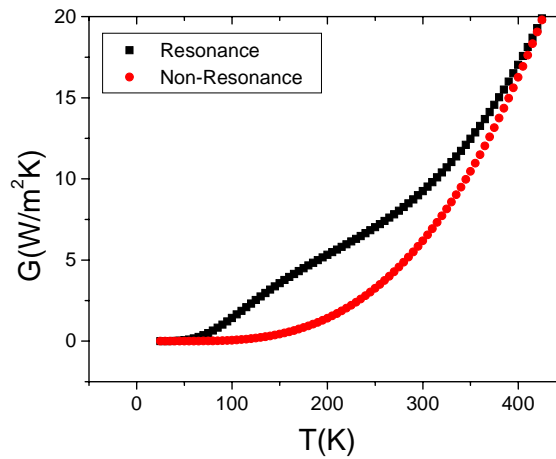
$$f(x, x_c, x_\parallel, x_\perp) \equiv \frac{x^2 e^x}{(e^x - 1)^2} \sqrt{x^2 - x_c^2 + x^2 \left( \frac{x_\perp^2 - x_\parallel^2}{x^2 - x_\perp^2} \right)}$$

Thin films = Planar waveguides = Planar microcavities

Anvar A. Zakhidov, University of Texas at Dallas



## Thin films: resulting enhancement of thermal conductivity (Again from difference in DOS)

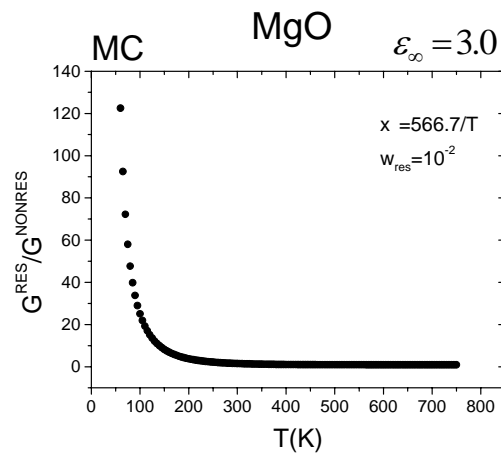


MgO

Anvar A. Zakhidov, University of Texas at Dallas



## Relative Enhancement of Heat Conductance by Polaritonic Effect in MICROCAVITY: The ratio of $G_{res}$ to $G_{nonres}$



Anvar A. Zakhidov, University of Texas at Dallas



## Thick samples: $\Lambda(\omega) < d$ Diffusive heat transfer



The thermal conductivity  $\kappa(T)$  can be calculated by the using of following well-known expression

$$\kappa(T) = \frac{1}{3} \sum_p \int C(\omega) v(\omega) \Lambda(\omega) d\omega, \quad (4)$$

where  $\omega$  is the polariton frequency,  $C(\omega)$  is its thermal capacity,  $v(\omega)$  is its the group velocity, and  $\Lambda(\omega)$  is its mean-free-path. The sum is carried out over two transverse polariton polarizations  $p$ .

Phonon-polariton in heat conduction

V. R. Coluci<sup>1,3</sup>, A. A. Zakhidov<sup>1</sup>, and V. M. Agranovich<sup>1,2\*</sup>

<sup>1</sup>NanoTech Institute and Department of Chemistry, University of Texas, Richardson, Texas 830688

<sup>2</sup> Institute of Spectroscopy, Russian Academy of Sciences, 142190 Troitsk, Moscow Region, Russia and

Anvar A. Zakhidov, University of Texas at Dallas



## Specific Heat $C(\omega)$



$$\begin{aligned} C(\omega) &= \frac{1}{V} \frac{dE}{dT} = \\ &= \frac{D(\omega)}{V} \frac{(\hbar\omega)^2 e^{\hbar\omega/k_B T}}{k_B T^2 (e^{\hbar\omega/k_B T} - 1)^2}. \end{aligned}$$

$$E(\omega, T) = \hbar\omega \frac{D(\omega)}{\exp(\hbar\omega/k_B T) - 1},$$

where the density of states  $D(\omega)$  is given by

$$\frac{D(\omega)}{V} = \frac{4\pi k^2 dk}{(2\pi)^3 d\omega}.$$

Anvar A. Zakhidov, University of Texas at Dallas



## Mean free path $\Lambda(\omega)$ of Phonon-Polaritons:



Since the intensity  $I$  is proportional to the squared electrical field we have

$$I \sim |E|^2 \sim e^{i2kz} = e^{i2(n' + in'')\omega z/c} \sim e^{-2n''\omega z/c} = e^{-z/\Lambda(\omega)} \quad \Lambda(\omega) = \frac{c}{2\omega n''(\omega)}.$$

Using the relation

$$\frac{k^2(\omega)c^2}{\omega^2} = (n' + in'')^2 = \varepsilon(\omega) = \varepsilon' + i\varepsilon'', \quad (11)$$

and assuming weak absorption ( $(n'')^2 \simeq 0$ ) one can obtain

$$\varepsilon'(\omega) = \varepsilon_\infty \left( 1 + \frac{(\omega_\parallel^2 - \omega_\perp^2)(\omega_\perp^2 - \omega^2)}{(\omega_\perp^2 - \omega^2)^2 + 4\Gamma^2\omega^2} \right), \quad (12)$$

$$\varepsilon''(\omega) = \varepsilon_\infty \left( \frac{2\Gamma\omega(\omega_\parallel^2 - \omega_\perp^2)}{(\omega_\perp^2 - \omega^2)^2 + 4\Gamma^2\omega^2} \right), \quad (13)$$

$$n'(\omega) = \sqrt{\varepsilon'(\omega)}, \quad n''(\omega) = \frac{\varepsilon''(\omega)}{2n'(\omega)}. \quad (14)$$

Anvar A. Zakhidov, University of Texas at Dallas



## Check of Approximation for $\Lambda(\omega)$ in MgO

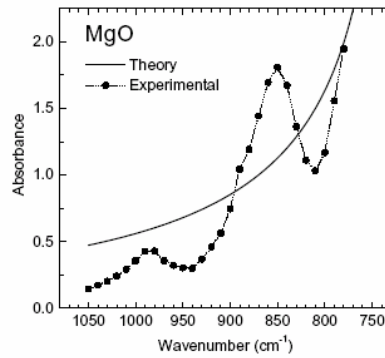


FIG. 2: Absorption spectra of MgO. Experimental points correspond to the MgO crystal at  $T=305$  K with  $0.16$  mm thick<sup>7</sup>. The dotted line just connects the points.

Anvar A. Zakhidov, University of Texas at Dallas



## Polaritonic K(T) in kinetic limit



$$\kappa(T) = \frac{k_B^3 T^2}{3\pi^2 \hbar^2 c} \left[ \int_0^{x_\perp} h(x) dx + \int_{x_\parallel}^{\infty} h(x) dx \right]$$

where

$$x(T) \equiv \frac{\hbar\omega}{k_B T}, \quad x_\perp(T) \equiv \frac{\hbar\omega_\perp}{k_B T}, \quad x_\parallel(T) \equiv \frac{\hbar\omega_\parallel}{k_B T},$$

$$h(x) \equiv \frac{x^3 e^x}{(e^x - 1)^2} \frac{\sqrt[3]{\varepsilon'(x)}}{\varepsilon''(x)}$$

Anvar A. Zakhidov, University of Texas at Dallas



## Comparison of Polaritonic contribution with experimental K(T)

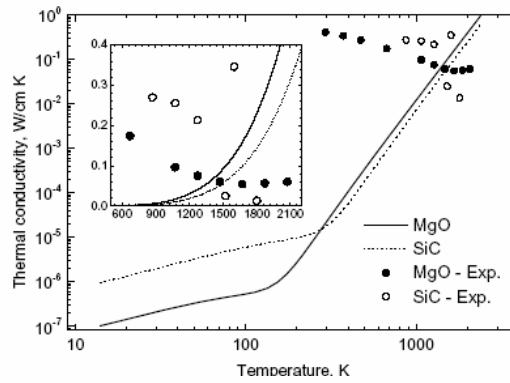


FIG. 3: Thermal conductivity as function of temperature. The lines were obtained using the expression (16). The inset graph is the zoom for the region  $600 \text{ K} < T < 2200 \text{ K}$ .

Anvar A. Zakhidov, University of Texas at Dallas



### Giant vibrational resonances in $A_6C_{60}$ compounds

Ke-Jian Fu,\* William L. Karney, Orville L. Chapman, Shiou-Mei Huang, Richard B. Kaner, Franck Diederich, Károly Holczér,<sup>†</sup> and Robert L. Whetten

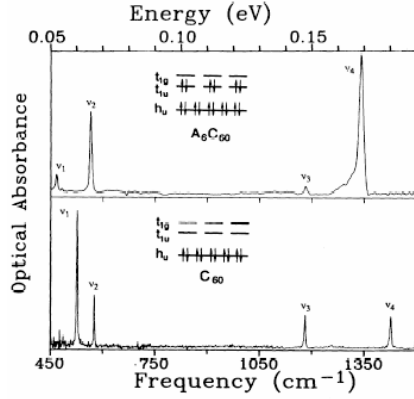


FIG. 1. Optical absorption spectrum in the midinfrared region of films of  $C_{60}$  (lower panel) and  $A_6C_{60}$  (upper panel), recorded

Anvar A. Zakhidov, University of Texas at Dallas

$F_{4u}$  optical IR mode is a great candidate for OP-Polariton with tunable K(T).

The intensity of optical absorption increase 88 times (!) upon doping  $x=6$  electrons in  $C_{60}$

Oscillator strength  $S \sim x^2$ , increases dramatically,  
Polariton Gap ( TO-LO splitting):  $\Delta \sim S^{1/2}$  ,  
So that the parameter  $\eta \sim x$

Polaritonic Thermal conductivity  $K(T) \sim \eta \sim x$ ,

K becomes tunable by doping level  $x$ .

K can be increased  $x$  times ( 6 in measured case)



Map of materials to choose  $\omega$  and  $\eta$

Material	$\omega_{\perp}$ (cm <sup>-1</sup> )	$\omega_{\parallel}$ (cm <sup>-1</sup> )	$\epsilon_{\infty}$	$\eta$
TlBr	48	114	5.41	1.375
TlCl	64	161	5.1	1.516
RbI	75	103	2.62	0.373
AgBr	81	136	4.62	0.679
AgCl	103	171	4.04	0.660
K-I	103	144	2.59	0.398
Kbr	116	168	2.34	0.448
NaI	117	181	3.03	0.547
RbCl	119	178	2.14	0.496
NaBr	135	210	2.63	0.556
KCl	144	216	2.16	0.500
RbF	160	293	1.93	0.831
NaCl	164	262	2.31	0.598
LiBr	173	354	3.16	1.046
KF	192	330	1.85	0.719
LiCl	204	425	2.75	1.083
NaF	246	424	1.72	0.724
LiF	304	660	1.9	1.171



$$\eta = \frac{\omega_{\parallel} - \omega_{\perp}}{\omega_{\perp}}$$

Anvar A. Zakhidov, University of Texas at Dallas



Map of  
materials  
to choose  
 $\omega$  and  $\eta$

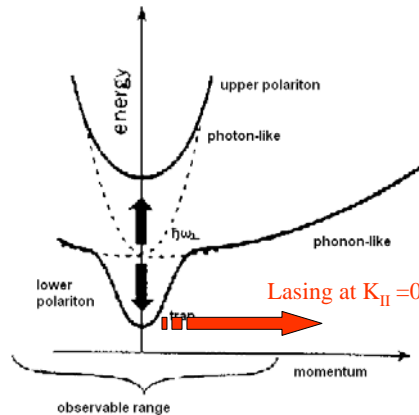
Material	$\omega_{\perp}$ (cm <sup>-1</sup> )	$\omega_{\parallel}$ (cm <sup>-1</sup> )	$\epsilon_{\infty}$	$\eta$
CdTe	140	171	7.2	0.221
CdSe	171	214	--	0.251
ZnTe	177	208	7.3	0.175
InSb	179	200	15.7	0.117
BaF <sub>2</sub>	184	336	2.16	0.826
ZnSe	203	252	5.9	0.241
CdTe	202	380	2.4	0.881
CuCl	172	210	3.71	0.221
InAs	210	243	12.3	0.157
SrF <sub>2</sub>	217	387	2.07	0.783
GaSb	225	233	14.4	0.036
CaF <sub>2</sub>	257	464	2.05	0.805
CdS	234	305	--	0.303
ZnS	274	349	5.2	0.274
GaP	366	402	9.1	0.098
MgO	394	719	2.956	0.825
SiC	796	970	6.52	0.219



Anvar A. Zakhidov, University of Texas at Dallas



## Idea of IR Phonon-Lasers: Condensation of phonon-polaritons in planar microcavities at Room-T



•Exciton-polariton condensation has been recently demonstrated in semiconductor MC.

•If condensation of lowest energy cavity phonon-polaritons is achievable, IR lasing would become possible with **zero pumping threshold**.

•The life-time of phonon-polaritons is a crucial parameter:

$t_{\text{lifetime}} > t_{\text{relaxation}}$   
is needed

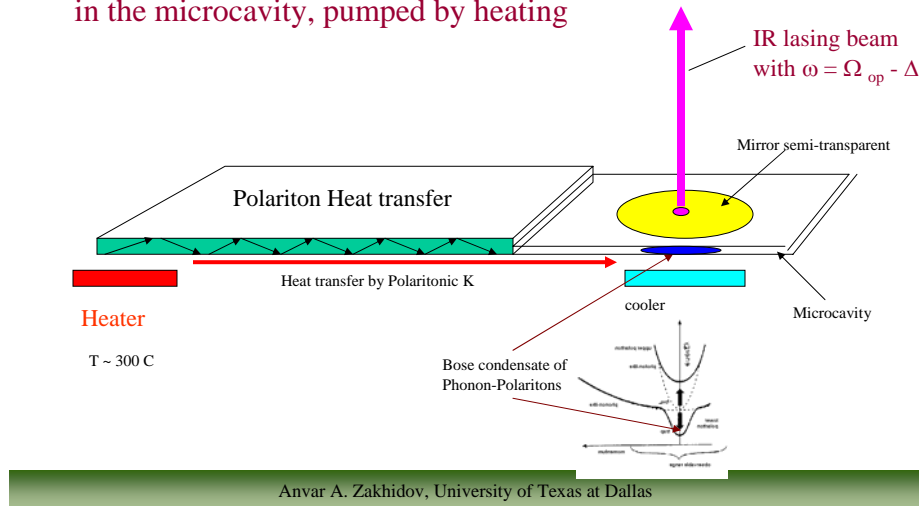
Anvar A. Zakhidov, University of Texas at Dallas



## New Concepts: “Phonon-Polariton Laser”



1. We propose: Phonon-Polariton laser in the microcavity, pumped by heating



## Phonon-Polariton Laser



- **Polariton Laser** - The polariton laser is a relatively new lasing mechanism postulated by several research groups and recently observed in GaAs at cryogenic temperatures by a Japanese researcher visiting in the US. The laser operates on the principle of Bose-Einstein condensation of excitonic polaritons within microcavities to align phase, with radiation from the condensate. Theoretical calculations by Professor Kavokin's group indicate that the same opto-electronic effects will be possible in wide-bandgap semiconductors at room temperature. Their calculations show that the kinetic blocking of polariton relaxation preventing formation of the B-E condensation of polariton phase at low temperatures should disappear at higher temperatures. These lasers have very low threshold currents, are very efficient, produce very little heat, and should have applications in very low power optical communications and optical computing.

Anvar A. Zakhidov, University of Texas at Dallas



## European Activities



- **New Laser Principle for Low Power and Fast Optoelectronic Devices:** Dr. Harvey, of ARL-ERO, visited Blaise Pascal University, FR, to discuss new room temperature lasing mechanism based on Bose-Einstein condensation in wide-bandgap semiconductor microcavities. This opens the door to very low power laser communications, THz optical signal processing, quantum computing, spintronic devices, THz modulation in photonic bandgap structures, and THz electronic signal processing. Such devices will enable the communication and processing of the massive amounts of data necessary to support FCS concepts. The Physics Dept at, Blaise Pascal University, has a very strong theoretical program and a good experimental program which is focusing on electron-light interactions in semiconductors, in particular excitonic polariton effects in semiconductor microcavities. Professor Kavokin leads a European collaboration funded by the European Community and focused on phenomena in semiconductor microcavities under the EC Framework program on “High Technology for Communications and Information Processing”.

Anvar A. Zakhidov, University of Texas at Dallas



## Polariton Transistor



- **THz Excitonic Polariton Transistor and THz Optoelectronic Devices** - The Blaise Pascal group has shown theoretically and experimentally that the excitonic polaritons can be accelerated in the plane of microcavities, with the gradient of the microcavity thickness acting as the forcing function. Observed velocities are one to two orders of magnitude faster than the electronic ballistic transport in bulk semiconductors, with the potential for using the polaritons as carriers in a very fast transistor type device and for ultrafast optical processing.

•

Anvar A. Zakhidov, University of Texas at Dallas



## Polaritonics: bridging the gap between electronics and photonics



- Between electronics and photonics there exists a frequency gap of approximately 2 octaves, i.e. the frequency range between 100~GHz and 10~THz. Here we demonstrate that **phonon-polaritons in ferroelectric crystals** like LiNbO<sub>3</sub> or LiTaO<sub>3</sub> may be able to bridge this gap. The ability to fabricate structures within the crystal by femtosecond laser machining facilitates all integrated signal guiding and processing. Spatiotemporal imaging is employed for direct visualization of the electromagnetic field within the crystal. Polaritonic resonators, waveguides, photonic crystals and focusing, dispersive, and diffractive elements will be demonstrated.
- **Authors:** David Ward, Thomas Feurer, Eric Stats, Joushua Vaughan, Keith Nelson, Massachusetts Institute of Technology)

Anvar A. Zakhidov, University of Texas at Dallas



## Conclusions on K(T) by Phonon-Polaritons



1. Ph-Polaritons are found to contribute to K(T) of thin films, with T-peak. Position of T-peak depends on  $\Omega_{op}$ , the line width of OP and the TO-LO splitting,
2. K(T) can be 10-20 times stronger than the conventional radiative contribution to K by free photons.
3. T-peak shifts to lowest T in microcavities ( $L \sim 1-10 \mu m$ ), which can be used in cryogenic heat transfer.
4. To create a material with high enough polaritonic K(T) at RT, compared to the usual, phonon K<sub>ph</sub> one should create an organic material with OP at 1500-2000 cm<sup>-1</sup>, which has large oscillator strength. In organic materials K<sub>ph</sub> is usually low (< 0.1-1 W/mK), the K<sub>pol</sub> can become a main contribution.
5. One candidate for polaritonic heat pipe, can be a doped fullerene film M<sub>x</sub>C<sub>60</sub> in which a giant oscillator strength S enhancement is found, which is quadratic in doping level x:  $S \sim x^2$ .
6. The strong dependence of K<sub>pol</sub>(T) on S(x) leads to tunability of K(T) by charge transfer and thus may be used in "polariton-transistors", in which K can be amplified by charging gate G.
7. Phonon-Polaritons can be used for "Polariton-lasers", which will emit monochromatic and coherent IR radiation, due to Bose-Einstein condensation in microcavity.

Anvar A. Zakhidov, University of Texas at Dallas



## Our Main Systems and Materials:



1. New Mechanism of Phonon-Polariton Thermal Conductivity

2. Carbon Nanotube Systems with Enhanced  $K(T)$  CNT:

- CNT Fibers and Yarns and
- Oriented CNT-ribbon aerogels

Anvar A. Zakhidov, University of Texas at Dallas



## PHONON TRANSISTOR in NANOTUBE FIBERS with Electrochemical Gate

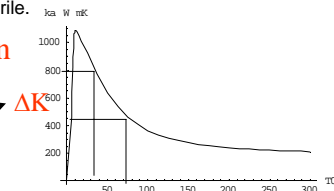


Capacitance of 134 F/gm, versus usual 15-30 F/gm.

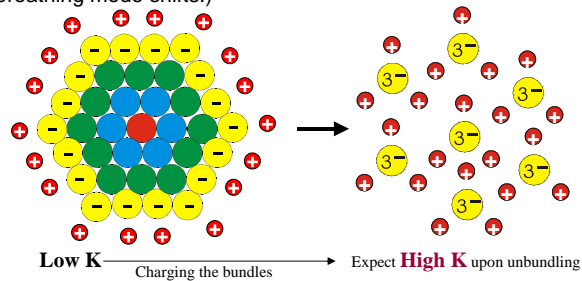
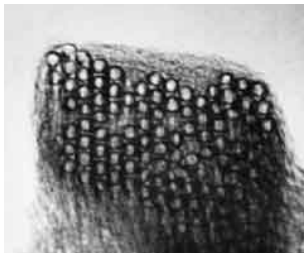
**Electrolyte:** 1.0 M tetrathylammonium hexafluorophosphate in acetonitrile.

Debundling should decrease tube-tube phonon scattering, and increase  $K(T)$  dramatically:

**Phonon Transistor**



Increased capacitance results from debundling.  
(Raman measurements of radial breathing mode shifts.)



Anvar A. Zakhidov, University of Texas at Dallas

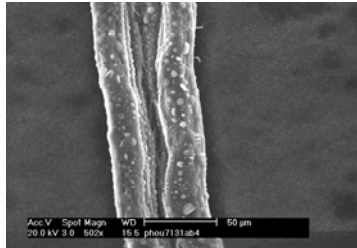


Prove of Unbundling: Twice Increased Diameter of Fibers



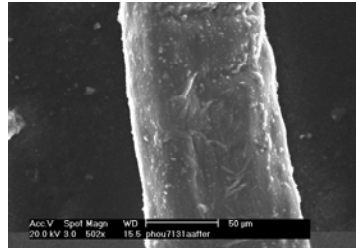
## SEM images of Purified Unannealed Oleum -spun HiPCo fibers

Phonon Transistor in 'OFF' state



Before cycling in EMII  
Fiber diameter  $\approx 50 \mu\text{m}$

Transistor in 'ON' state



After cycling in EMII  
Fiber diameter  $\approx 100 \mu\text{m}$

Anvar A. Zakhidov, University of Texas at Dallas

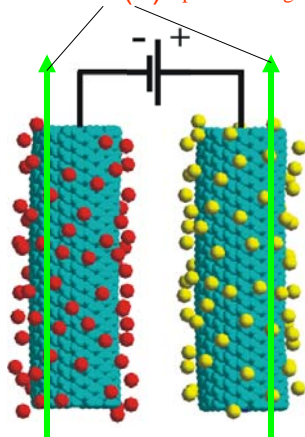
51



## Phonon Transistor with Electrochemical Charge Injection Gate



Increased  $K(T)$  upon G voltage



Gate function:

$C = \text{charge/voltage}$   
 $C = \text{Area} \times \text{dielectric constant} / d$   
*Area/weight is above  $300 \text{ m}^2/\text{gm}$ ;  
 $d$  is in nanometers*

Modulation of Thermal Conductivity  $K(T)$

*Charge injection will cause change in tube-tube interaction, which changes  $\Omega$  of intertube phonon and modulate The Tube-Tube scattering.*

Anvar A. Zakhidov, University of Texas at Dallas



## Charging Setup:

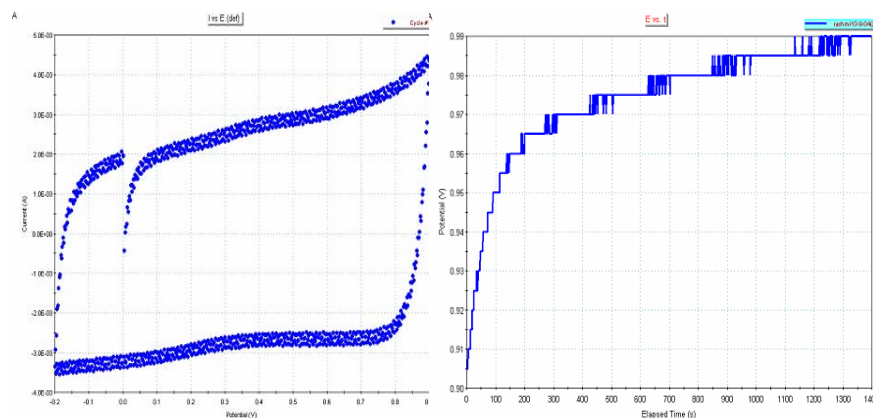


- SWNT paper was used to charge and study the effect of charge injection in field emission characteristics. SWNT paper was used as both the positive and negative electrode.
- Chronopotentiometry was used for performing the double layer charging. Princeton model 273A instrument contains both a potentiostat and a galvanostat, and hence can perform both controlled potential (potentiostatic) and controlled current (galvanostatic) experiments.
- Cyclic Voltammetry was also performed before the charging by cycling from  $-0.2$  to  $0.9\text{V}$ . The direction of the potential is reversed at the end of the first scan. This has the advantage that the product of the electron transfer reaction that occurred in the forward scan can be probed again in the reverse scan. It is a powerful tool for the determination of formal redox potentials, detection of chemical reactions that precede or follow the electrochemical reaction and evaluation of electron transfer kinetics.

Anvar A. Zakhidov, University of Texas at Dallas



## Charging I-t Curves

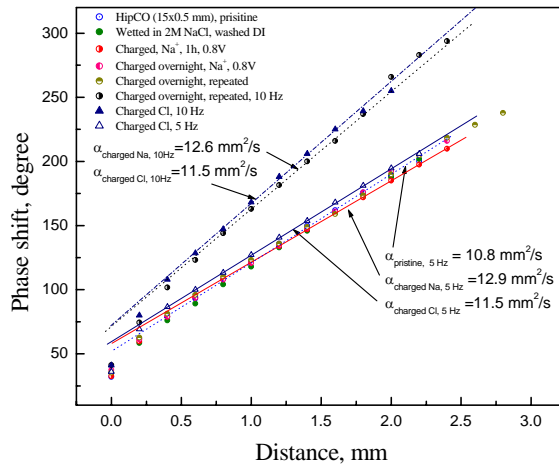


Capacitance of SWCNT paper is very large:  $C \sim 20\text{-}30\text{ F/g}$

Anvar A. Zakhidov, University of Texas at Dallas



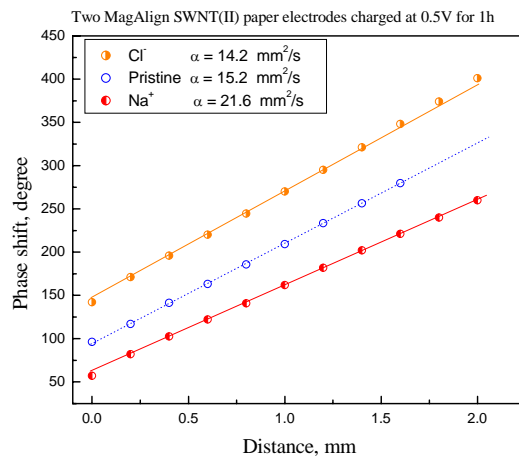
## Tuning K by Charge Injection



Anvar A. Zakhidov, University of Texas at Dallas



## Tuning K by Charge Injection: A Step towards Phonon Transistor



Thermal diffusivity and thus thermal Conductivity of CNT is proved to be tuned by Charge injection into

Oriented CNT paper at  $V = 0.5 \text{ V}$

Effect is 25 %:

From  $D = 15.2 \text{ mm}^2/\text{s}$  to  $21.6 \text{ mm}^2/\text{s}$

Anvar A. Zakhidov, University of Texas at Dallas

## Draw and Twist of yarns from MWNT forests

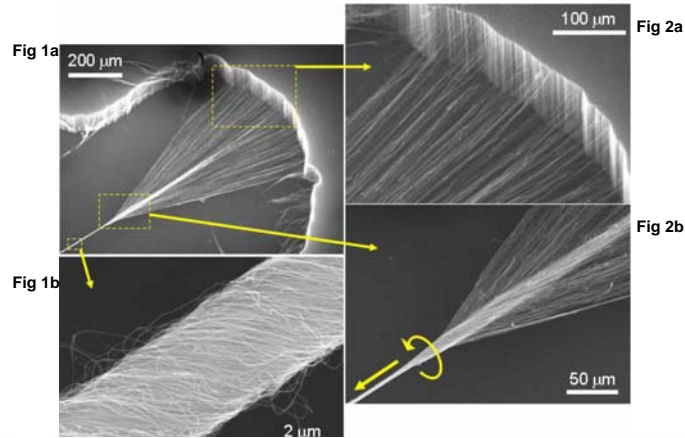
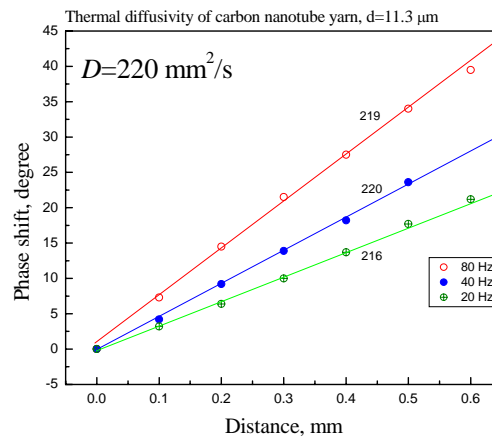


Fig. S2. SEM micrographs showing the structures formed during the draw-twist process. The relationships between the SEM micrographs of Fig. 1 and Fig. 2A are shown, as well as a higher magnification image of the partially bundled MWNTs being pulled from the forest wall. The draw twist process was interrupted, and the sample was transferred to a SEM for recording these images.

## High Thermal Diffusivity of Single CNT Yarn: Better than Cu



Anvar A. Zakhidov, University of Texas at Dallas



## Thermal conductivity of Single CNT Yarn: Better than Cu



- The thermal conductivity  $\lambda$  of single-strand MWNT yarn was obtained at room temperature from the relationship  $\lambda = \rho C_p D$ , by measuring the thermal diffusivity  $D$ , density  $\rho$ , and specific heat capacity  $C_p$ . The measurements of  $D$  were carried out using the laser flash technique.
- One end of a specimen of length  $L$  is uniformly irradiated by a laser beam  $Q = Q_0 \sin \omega t$ .
- 
- $\lambda = \rho C_p D = 0.8 \text{ g/cm}^3 \cdot 0.715 \text{ J/gK} \cdot 2.20 \text{ cm}^2/\text{s} = 1.25 \text{ W/cm} \cdot \text{K} = 125 \text{ W/m} \cdot \text{K}$ ,
- where the specific heat capacity of graphite with density  $2.26 \text{ g/cm}^3$ :  $C_p(300\text{K}) = 8.58 \text{ J/(mol K)} = 0.715 \text{ J/gK}$  [1], (For comparison the specific heat capacity value for  $10 \mu\text{m}$  Amoco P-55 carbon fibers, with density of  $2 \text{ g/cm}^3$  at  $25^\circ\text{C}$  is  $0.717 \text{ J/gK}$  [2]),  $\rho = 0.8 \text{ g/cm}^3$  is the density of fiber, and  $D = 2.20 \text{ cm}^2/\text{s}$  is the thermal diffusivity of the fiber.
- For comparison, the thermal diffusivity of thin wire ( $100 \mu\text{m}$ ) of copper and gold are much lower:  $D_{\text{copper}} = 117 \text{ mm}^2/\text{s}$ ,  $D_{\text{gold}} = 130 \text{ mm}^2/\text{s}$ .

Anvar A. Zakhidov, University of Texas at Dallas



## 'Draw-Twist' process to convert MWNT in a forest to 'Twisted Yarns'



### Multifunctional Carbon Nanotube Yarns by Downsizing an Ancient Technology

M. Zhang, Ken Atkinson, Ray Baughman, *Science* 306 (2004) 1358

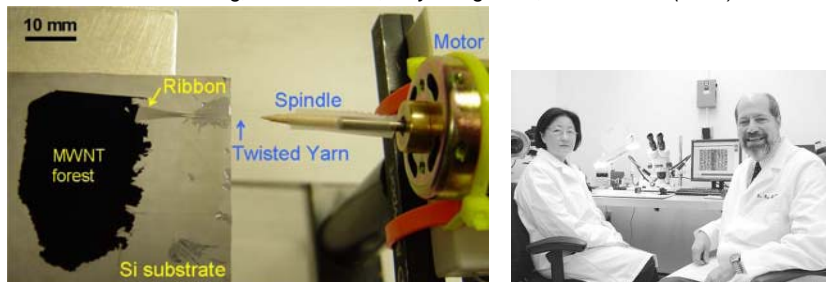


Fig. S1. Photograph taken during interruption of the draw-twist process used to convert MWNTs in a forest to a twisted MWNT yarn. The overlapping images of both the nanotube wedge and yarn are a result of reflection in the silicon substrate.

Anvar A. Zakhidov, University of Texas at Dallas



## SEM images of 'Twisted Yarns'

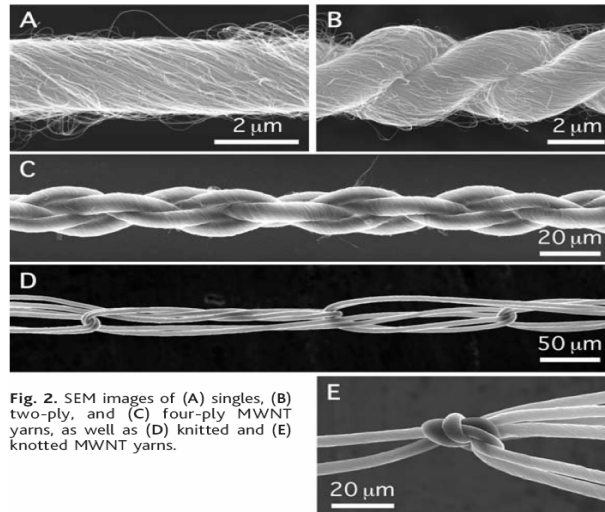
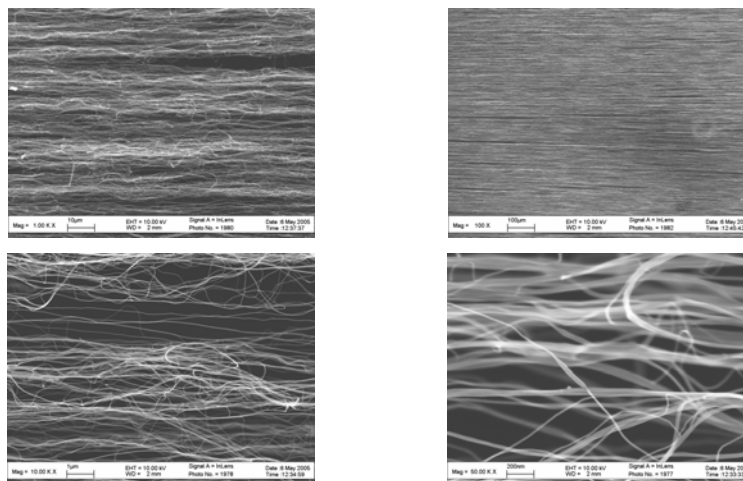


Fig. 2. SEM images of (A) singles, (B) two-ply, and (C) four-ply MWNT yarns, as well as (D) knitted and (E) knotted MWNT yarns.

Anvar A. Zakhidov, University of Texas at Dallas



## Oriented Multiwall CNT Thin Sheets: Aerogels with strongly anisotropic K(T)



Anvar A. Zakhidov, University of Texas at Dallas



## NanoTech Institute MWNT Sheet Fabrication Process



Sheets (presently 5 cm X 1 m) are fabricated At 3 m/minute. These width and length are not fundamentally limited and the rate is limited by our present draw apparatus.

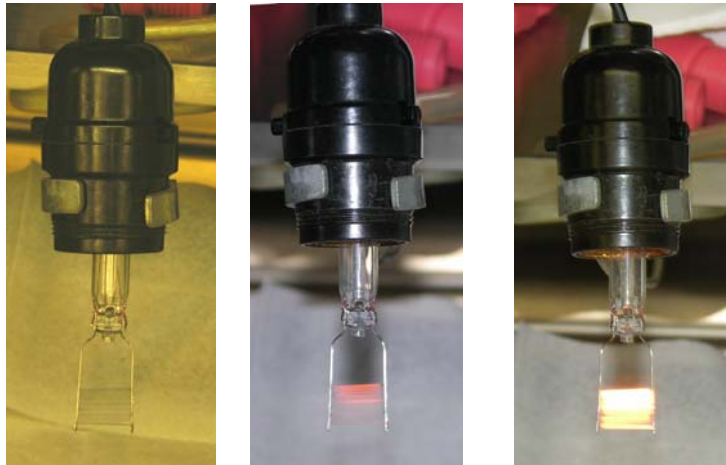
We have promising initial results for diverse applications:

- \* transparent elastomeric electrodes;
- \* light-emitting diodes;
- \* incandescent sources of polarized light;
- \* two-dimensionally reinforced composites; & microwave absorbing appliqué

Anvar A. Zakhidov, University of Texas at Dallas



## Spun nanotube sheets as an incandescent light source.

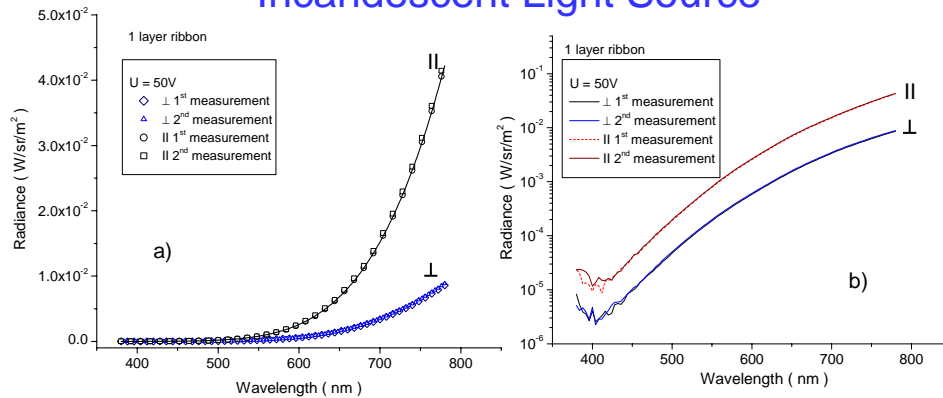


The light output is polarized, with a degree of polarization that increases with wave length from 0.6 at 500 nm to 0.66 at 780 nm.

Anvar A. Zakhidov, University of Texas at Dallas



## Polarized Emission of Nanotube Sheet Incandescent Light Source

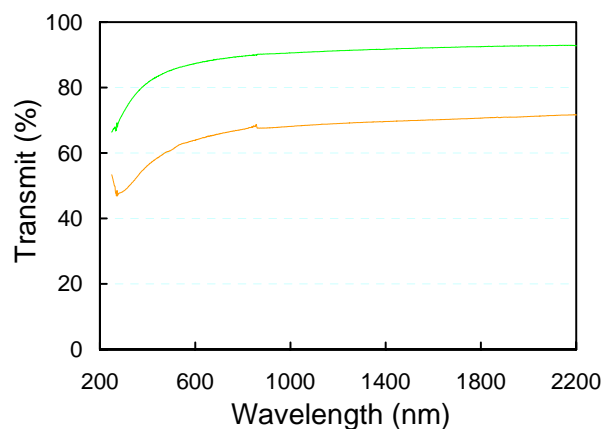


Spectral radiance of incandescent light from single sheet of parallel carbon nanotubes: a) linear scale, b) semi logarithmic scale. The solid line in a) is a fit by black body radiation with  $T=1350$  K.

Anvar A. Zakhidov, University of Texas at Dallas



## Optical Transparency of MWNT Sheet



Polarizer Alignment:  
*Perpendicular*  
*Parallel*

Resistance:  $\sim 600 \Omega/\square$  (in aligned direction)  
 $\sim 15 \text{ K}\Omega/\square$  (in cross direction)

Anvar A. Zakhidov, University of Texas at Dallas